



APPENDIX A

"CLEAN" VERSION OF EACH PARAGRAPH/SECTION/CLAIM

37 C.F.R. § 1.121(b)(ii) AND (c)(i)

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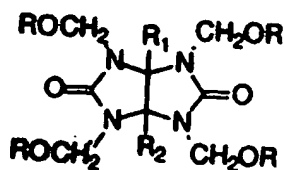
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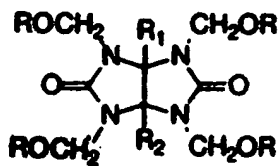
A

Crosslinkers for the compositions of this invention are Glycoluril derivatives of general formula:



Glycouril derivatives

in which R is -CH₃, and R₁ and R₂ can individually be selected from alkyl groups having 1 -6 carbon atoms exemplified by (a) tetrakis-methoxymethyl-3 α ,6 α -diethylglycouril,

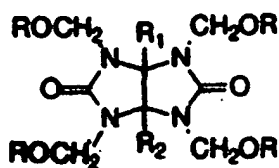


Glycouril derivatives

(b) tetrakis-methoxymethyl- 3 α -methyl-6 α -propylglycoluril and

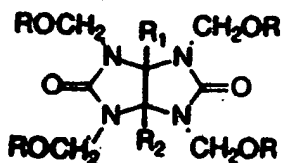
(c) tetrakis-methoxymethyl- 3 α -methyl-6 α -butylglycoluril as shown in the following formula:

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Glycouril derivatives

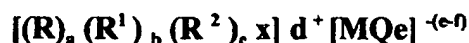
Al R¹ and R² can also be selected from alkyl and aryl group or hydrogen and aryl groups like (d) tetrakis-methoxymethyl-3 α -phenylglycouril and (e) tetrakis-methoxymethyl-3 α -methyl-6 α -phenylglycoluril as shown in the following formula:



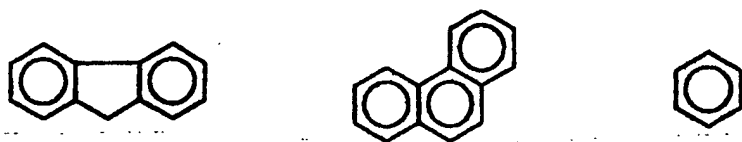
Glycouril derivatives

The composition of the present invention also preferably includes a photoacid generator (PAG) which can be selected from onium salts of Group IV elements as disclosed in U.S. Patent No, 4,175,972 the teaching of which is incorporated herein by reference and aromatic onium salts of Group Va elements disclosed in U.S. Patent No. 4,069,055

the teaching of which is incorporated herein by reference. Aromatic Group IVa onium salts include those represented by formula:

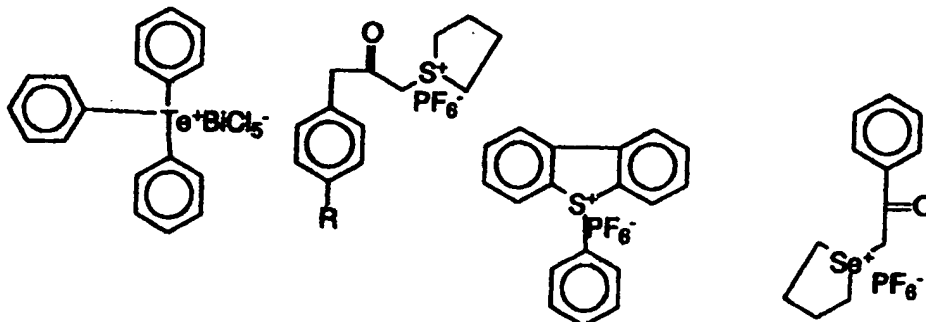


where R is a monovalent aromatic organic radical, R¹ is a monovalent organic aliphatic radical selected from alkyl, cycloalkyl and substituted alkyl, R² is a polyvalent organic radical forming a heterocyclic or fused ring structure; x is a Group IVa element or metalloid, Q is a halogen radical, a is a whole number from 0 to 3 inclusive, b is a whole number from 0 to 2 inclusive and c is a whole number equal to 0 or 1. Radicals included by R are, for example, (C₆ - C₁₄) aromatic hydrocarbon radicals such as phenyl, tolyl, xylyl, naphthyl, anthryl and radicals substituted with up to 1 to 4 monovalent radicals, such as C(1-8) alkyl, C(1-8) alkoxy, nitro, chloro, fluoro, and hydroxy; arylacyl radicals such as phenylacetyl; aromatic heterocyclic radicals such as pyridyl and furyl; R¹ radicals include C(1-8) alkyl, substituted alkyl such as -C₂H₄OCH₃, -CH₂-COCH₃, etc.; R² radicals include structures such as:

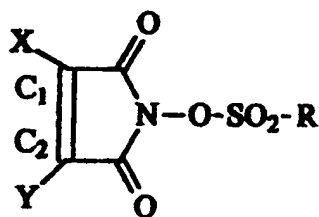


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Complex anions included by $\text{MQe}^{(+)}$ of above formula are, for example : BF_4^- , PF_6^- , SbF_6^- , FeCl_4^- , SnCl_6^- , SbCl_6^- , BiCl_6^- , AlF_6^{3-} , GaCl_4^- , InF_4^- , etc. Group VI a onium salts included by above formula are, for example:



Among non-metallic radiation degradable acid generators suitable for use in compositions of this invention are N-sulfonyloxyimides of the form:



where R is selected from the group consisting of toluene, benzene, CF_3 , CF_2CF_3 , $-(\text{CF}_2)_n-\text{Z}$ where $n = 1$ to 4 and Z is H or alkyl or aryl and where X and Y either (1) form polycyclic ring which may or may not contain heteroatoms, or (2) form a fused aromatic

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a¹
ring or (3) may be independently H, alkyl or aryl group and C₁ and C₂ may form a single or double bond. The compositions of the present invention are preferably carried in a solvent or solvent system. The solvent or solvent system used in carrying these compositions preferably fulfills the need for providing uniform coating with complete coverage of the resist components on the substrate. The solvent preferably evaporates at temperature such that the photoactivity of the resist is not adversely impacted upon a drying step and will not take part in the operation of the photoresist (to be inert toward the phenolic resin and/or crosslinker). Typical of these solvents are propyleneglycol monomethyl ether acetate {PGMEA}, ethyl-3-ethoxypropionate {EEP}, methoxypropanol, ethoxypropanol, butoxypropanol and ethyl lactate.

For formation of the negative tone image, the composition of the present invention is applied (for example, by spin-coating) on a substrate and the remaining solvent is evaporated by heating the substrate on a hot plate to temperatures of about 70-130° C for 1 -3 minutes. The film is then imagewise exposed to E-beam radiation at 100 kV. 200-370 nm upon which the radiation degradable component of the composition degrades and forms catalytic amount of acid. The crosslinking reaction between aliphatic hydroxyl calixarene dendrimer shown above and crosslinker is then accelerated by heating the substrate from 70-120° C for preferably one to three minutes. The images are thus rendered less soluble or insoluble in organic solvent. Typical of these solvents are propyleneglycol monomethyl ether acetate (PGMEA), ethyl-3-ethoxypropionate (EEP), methoxypropanol, ethoxypropanol, butoxypropanol,

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and ethyl lactate, and in the developing stage, the unexposed areas are removed by these solvents.

In another embodiment of this invention, the glycoluril derivatives which are used as crosslinkers are synthesized from corresponding dicarbonyl compounds as described.

A Glycoluril derivatives of this invention are highly soluble in the organic solvent of choice, and compared with unsubstituted glycoluril. For purposes of explication of the present invention, the terms "glycouril" and "glycoluril" connote the identical compound. The expression "organic base" represents a compound having basic (alkaline) properties.

General procedure for preparation of alkyl-substituted Glycourils

Synthesis of alkyl-substituted glycolurils is exemplified by the synthesis of 3 α -methyl-6 α -propylglycoluril described below.

2,3-hexandione (1.0 mole) and urea (3.0 mole) were added to 2.0 liters of 2% hydrochloric acid and the solution is stirred at room temperature for 24 hours (alternatively, the solution could be refluxed for 2-3 hours). The precipitate is filtered and washed with water and then ethanol and dried. The off-white powder is then crystallized in acetic acid to give analytically pure titer compound in 45% yield.

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